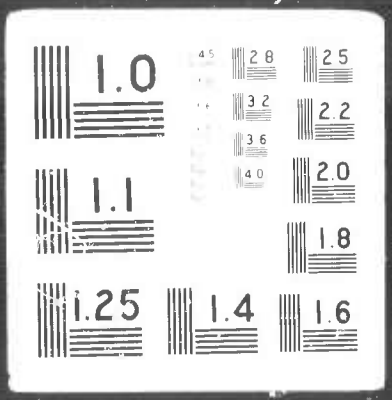

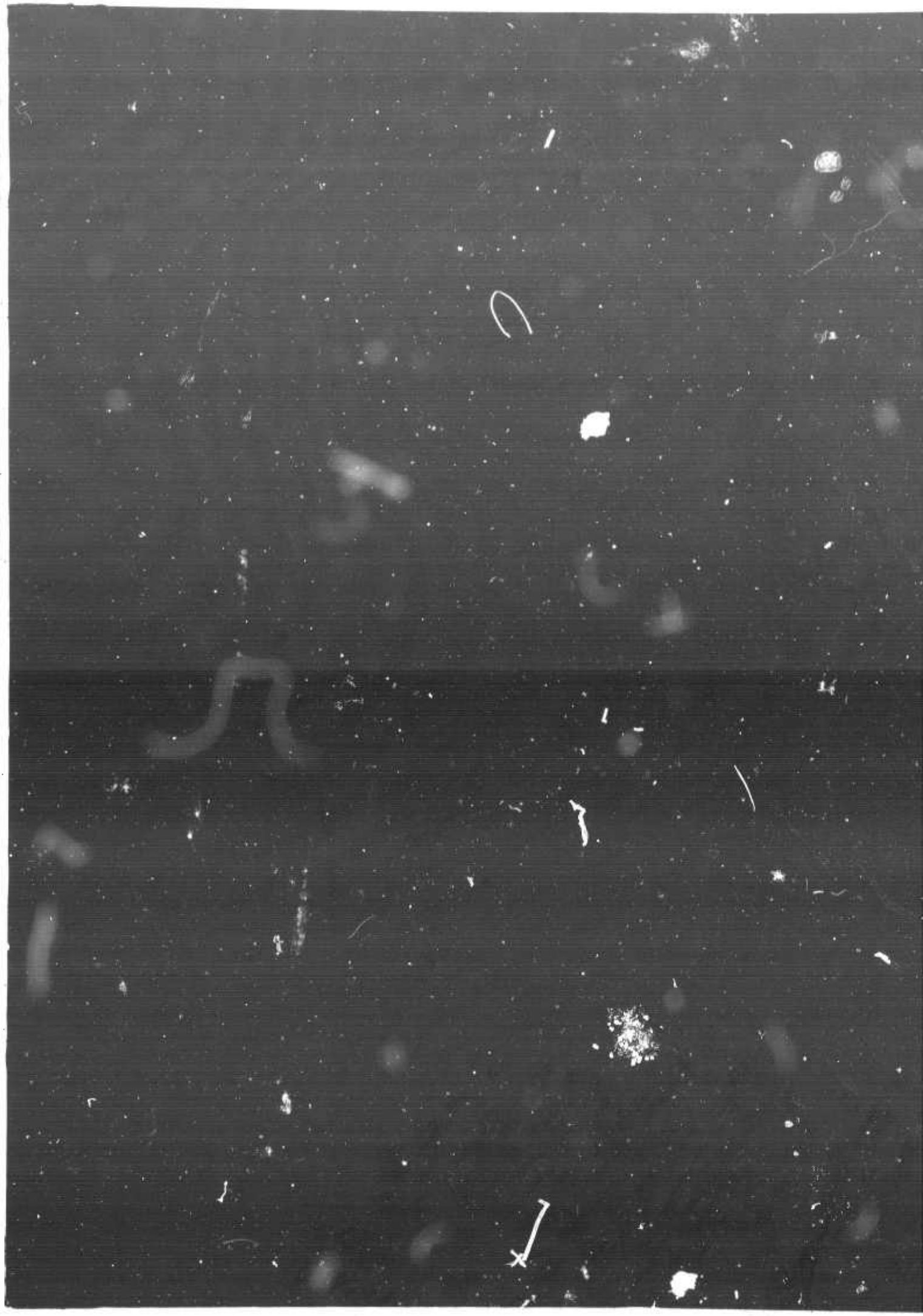

SPECTRAL PROPERTIES OF PASSIVELY Q-SPOILED LASERS

SemiAnnual Technical Summary Report

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ABSTRACT

Picosecond pulse outputs, tunable over a broad spectral range, have been demonstrated in an organic dye laser.

FOREWORD

This report was prepared by the Optical Physics Division of Korad Department, Electronics Division, Union Carbide Corporation, Santa Monica, California, under Contract Nonr-5150(00) entitled "Spectral Properties of Passively Q-Spoiled Lasers". The study was conducted under the project leadership of B. H. Soffer with J. W. Linn participating.

Section 1

INTRODUCTION

The object of this program is an experimental research study of the spectral properties of passively Q-spoiled lasers and the properties of organic dye lasers. This study is directed toward a further understanding of the spectral and physical behavior of these laser and Q-spoiling materials.

In the previous SemiAnnual Technical Summary Report we reported work on the effects of incorporating passive bleachable absorbers into the organic dye laser in order to achieve mode locking. During this period we have successfully achieved the mode locking of the organic dye laser and report the observation of the picosecond pulse structure. Furthermore, picosecond pulse outputs tunable over a broad spectral range are described.

Section 2

TECHNICAL DISCUSSION

A. Stimulated Emission in Organic Dyes

1. Mode Locking

In our quarterly report (1 Oct - 31 Dec 1967) we reported preliminary observations which indicated a degree of mode locking in an organic dye laser which might be sufficient to yield picosecond pulses. A Rhodamine 6G dye laser was pumped with the second harmonic of a mode-locked Nd laser. The length of the organic dye laser was made equal to or a submultiple of the pump cavity length, and trains of pulses were observed with subnanosecond resolution in the output of the dye laser. The existence of these pulses, with proper temporal periodicity is a necessary condition for the dye laser to be mode locked to some degree. Similar observations done independently and simultaneously by another group were recently reported in the literature.[1]

A mode-locked neodymium glass laser was constructed for use as a pumping source for the organic dye laser. A photo of the experimental apparatus and a schematic diagram may be seen in Figures 1 and 2. All intracavity reflective elements were set at the Brewster angle and mode locking was effected by the use of a bleachable absorber near the 100% reflector in a cell positioned to minimize the intensity of satellite spikes on the main pulse train. The pulse was

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[1] W. H. Glenn, M. J. Brienza and A. J. DeMarice, Appl. Phys. Letters 12, 54 (1968)

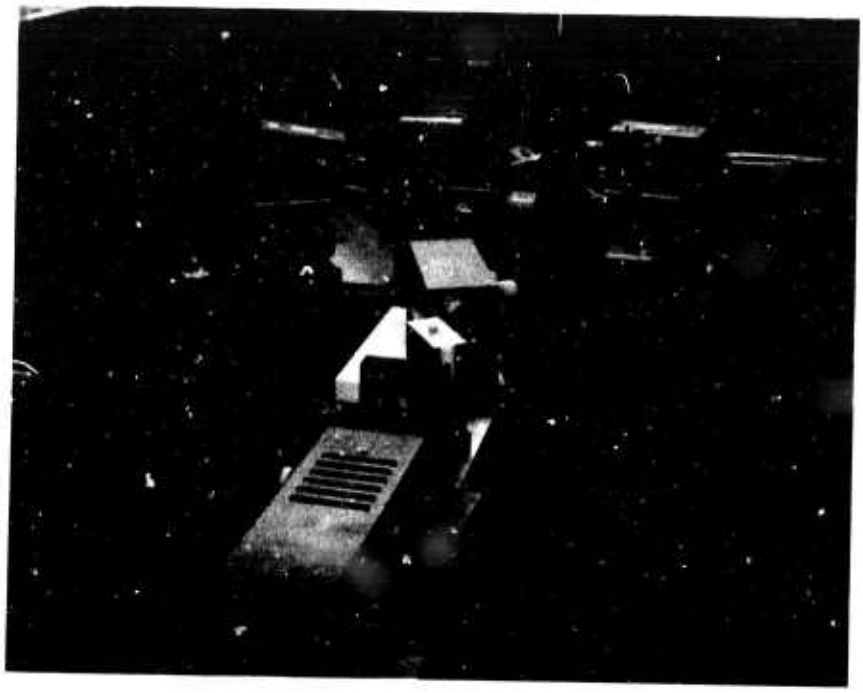

Figure 1

Photograph of Experimental Apparatus

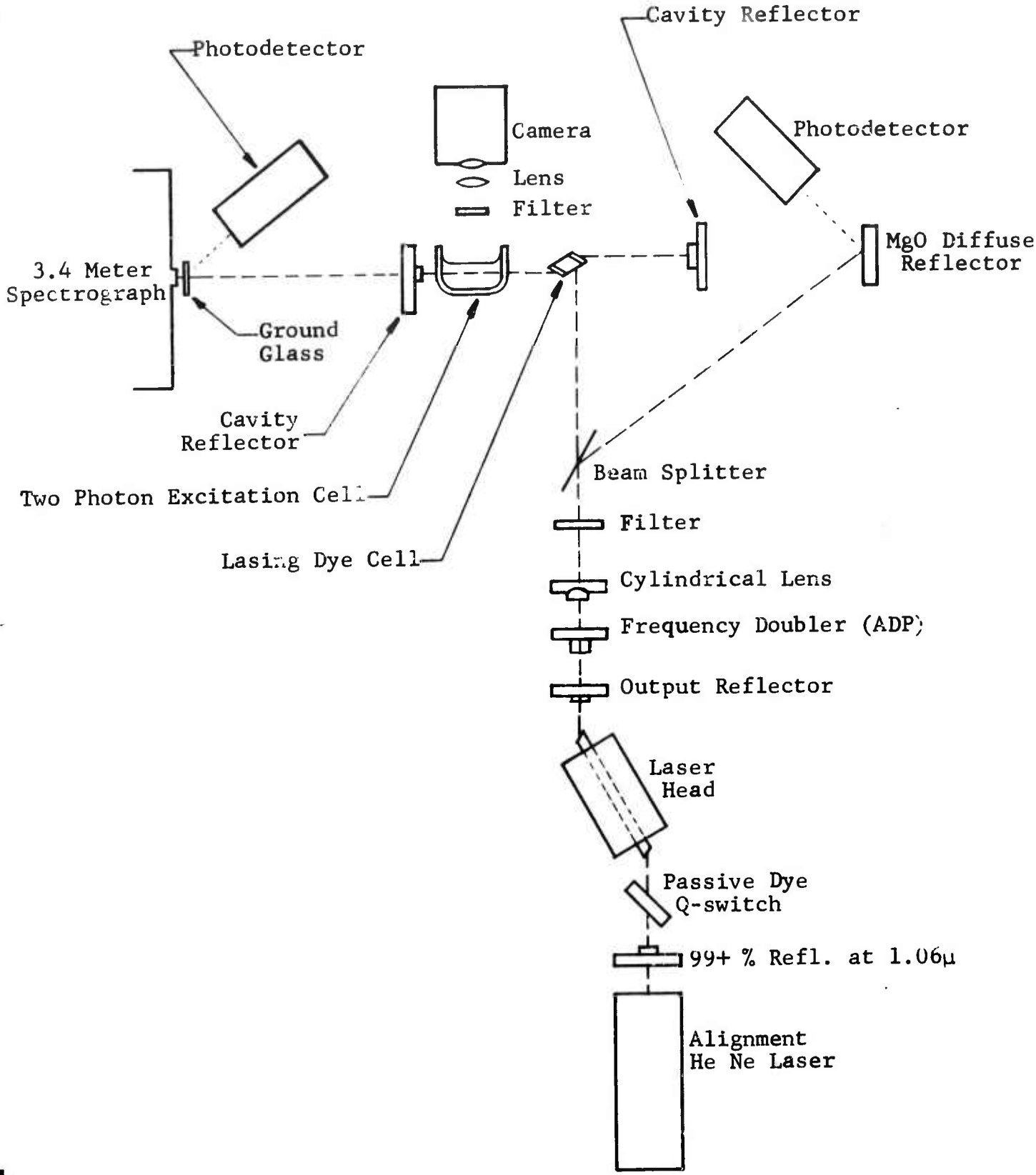

Figure 2

Schematic Diagram of Experimental Apparatus

structured in groups of 10 or 11 individual pulses separated by 1×10^{-10} sec, an interval determined by the internal round trip in the cavity output reflector. The surfaces of this glass reflector were wedged at 2' and one surface was coated with a 30% dielectric reflector creating a lower resonator. These groups of pulses themselves, were separated by 3×10^{-9} sec, 2L/C of the cavity with typically 10-15 sets between the half power points. Picosecond pulses were detected by the two photon excitation of fluorescence technique. A photo of this representation of the pulses is shown in Figure 3. A solution of Rhodamine 6G in ethyl alcohol was employed as the fluorescent medium. The second harmonic of this laser generated by an ADP crystal revealed the same pattern of pulses with the same temporal duration of $4 \pm 1 \times 10^{-12}$ sec. A solution of Esculin in ethyl alcohol was employed in this case as the two photon fluorescent medium. The spectral bandwidth of the output was 150 cm^{-1}.

These second harmonic pulses were used to transversely pump a Rhodamine 6G dye laser. An ethanolic solution of the dye with decadic absorption coefficient equal to 10 at 530 nm was contained in a cell constructed with internal and external window surfaces disposed at their respective Brewster angles. See Figures 1 and 2. The internal length of the cell was 1.15 cm. Laser action in unwanted directions was suppressed by degrading all optical surfaces excepting the windows. Wedged dielectric coated mirrors of ~ 99% reflectivity and of 60-90% output reflectivity were employed.

When the dye laser cavity optical length L_ℓ, was made equal to (or to a small submultiple of) the pump cavity optical length L_p and the Rhodamine laser dye cell put at the end of the cavity, trains of fully modulated pulses of periods equal

Figure 3

Two Photon Fluorescence Representation of Picosecond Pulses

to (or to small submultiples of) the pump pulse were observed with instrumental temporal halfwidth of 7×10^{-10} sec using a planar photodiode and traveling wave oscilloscope. With $L_p = L_\ell$ and the laser cell in intermediate harmonic positions L/n, n = 3, 4; two pulses separated by 2L/nc occurred for each pumping pulse with one predominantly intense, as may be expected if one considers that pulses traveling in opposite directions in the cavity, reach the dye cell together only once per pump pulse. With the cell centered and $L_\ell = mL_p/n$; m/n = 2, 1, 2/3, 1/2; 2n/m pulses are obtained for each pumping pulse, spaced at 3. 3/2, 1, and 3/4 nanoseconds respectively. These results are an extension of findings reported earlier. When the pulse interval approaches the pulse resolution time, the overlap of the instrumentally broadened pulses reduces the apparent modulation drastically. When two or more pulses per pump pulse occur, the first two are of approximately equal intensity and any remaining ones show rapidly diminishing intensity but with larger statistical variations in decay rates on successive pictures. The fluorescent lifetime of Rhodamine 6G has been recently redetermined as 5.5×10^{-9} sec for an approximately equal concentration solution. This time is greater than any of the periods described above, a condition which would diminish the effectiveness of this dye were it used as a passively bleachable absorber to engender mode locking with the cavity dimensions and configurations described above. However, this is a driven system with periodic gain modulation. The superradiant lifetime over the pumping intensity range employed was found to be greater than 1×10^{-9} and less than 3×10^{-9} sec by removing the cavity reflectors and observing the ability of the emitted radiation to follow stimulation of diminishing periodicity. The effective cavity lifetime including the driven dye laser cell was inferred from the multipulse decay rates to be between 1 and 2 nsec.

2. Picosecond Pulse Outputs

The picosecond pulse structure of the dye laser was observed using the alcoholic solution of Esculin. Multiple exposures were often required and so for convenience the two photon fluorescence cells were sometimes positioned in the cavity, a procedure empirically justified by the finding of no significant change in the observed pulse shapes. More dispersive, scattering, and generally non-linearly behaving solvents then ethanol might be expected to cause complications. Several of the cavity configuration mentioned above were tried all yielding similar picosecond pulses patterns. The additional expediency of increasing the f# of the camera lens with inexpensive positive portrait lenses allowed the use of less sensitive but higher gamma emulsions (Polaroid type 52, $\gamma \sim 3.5$ and type 146 L, $\gamma \sim 2.5$) to photograph the two photon excited fluorescence. This is an aid in densitometrically determining the contrast ratio (see below), as the fractional error in contrast ratio, for an error in optical density, is inversely proportional to gamma.

The structure of the picosecond pulses closely followed that observed for the pump except that the pulse width was $1.1 \pm 0.1 \times 10^{-11}$ sec. The contrast ratio was determined to be 2 ± 0.2, and the contrast ratio of the pump pulse was found to be 1.8 ± 0.2. Several approaches have been taken to attempt to realize more of the full mode locking potential of the ~ 150 cm^{-1} dye laser bandwidth which might yield pulses as short as 2×10^{-13} sec in duration. However, no significance changes in pulse widths were noted when the spatial width of the pumping pulse was narrowed from 12 to 1 mm by focusing with a cylindrical lens in order to bound the 10^{-11} sec pulse length, when fine variations were made in the position of the

pulse in the dye cell and the position of the cell itself, or when operating the dye laser from 10 to as much as 20 times above threshold. Higher output mirror reflectivities produced only trivially shorter pulses within the estimated deviation.

One condition that might contribute to the pulse width is a frequency sweeping of the laser output, a circumstance reported for certain cyanine dye lasers. It should also be noted that because of the pulse structure of each laser burst in these experiments, each of the picosecond pulses displayed in the two photon fluorescence technique is actually an average over $> 10^2$ discrete pulses. Each pulse may be narrower than the average but broadened by aperiodicity within each 10^{-9} sec group of pulses or from one such 10^{-9} sec pulse to the next. Furthermore, as has recently been discussed, two photon fluorescence pictures of picosecond pulses with contrast ratios of less than three, the maximum value which can be obtained as for example in the case of fully mode locked operation, are fraught with uncertainties about the actual nature of the pulses. Contrast ratios of two which are observed here might indicate a free running oscillation or random (initial condition) mode locked laser, but certainly not necessarily. It should further be noted that all experimental problems as for examples, scattering, and imperfect overlap of the beams, would tend to reduce the observed contrast ratio. Because of this uncertainty we can quote only the bounds of the average peak power between a maximum and a minimum were there no picosecond pulse content: for the pump $8 \times 10^6 <$ P watts $< 10^8$; for the dye laser $2 \times 10^6 <$ P watts $< 2 \times 10^7$ using an output reflectivity of 60%. A smaller number of picosecond pulses might be expected

to yield higher powers. The full beam angle at half power density was measured as 2.0 mrad.

3. Tunable Picosecond Pulse Outputs

Efficiently confining the spectral output of the dye laser to 6 cm^{-1}, by replacing one of the cavity mirrors by a diffraction grating as described in an earlier publication, we have demonstrated a continuously tunable source of $1.1 \pm 0.1 \times 10^{-11}$ sec pulses over a half power tuning range of 600 cm^{-1}. Narrowing the spectral output much beyond this would, of course, broaden the pulse widths. The pulse pattern again resembled the pumping pulse structure and had a measured contrast ratio of $2 \pm .2$. Using a grating of 2,160 lines/mm of approximately constant first order reflectivity of 58% in the range of interest in the Littrow position and a 100% reflector, the average peak power output in the zeroth order at the peak of the tuning curve was greater than 50% of that obtained with the 60% dielectric reflector replacing the grating. The full beam angle at half power density was found to be 4.8 mrad.

Section 3

PLANS FOR NEXT PERIOD

This technique, providing tunable short pulses, is expected to work well with many families of dyes, both in liquid and plastic hosts covering the entire range from the near ultraviolet to the near infrared. The flash lamp pumped self mode locked dye laser would also be ameanable to this technique. Longitudinal pumping of the dye cell should, as has been demonstrated, reduces the dye laser beam divergence significantly. Single tunable picosecond pulses or pulses with other intervals should also be obtainable. We are continuing to investigate these possibilities.

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13. ABSTRACT

Picosecond pulse outputs, tunable over a broad spectral range, have been demonstrated in an organic dye laser.

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